Quantum transport efficiency and Fourier's law

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We analyze the steady-state energy transfer in a chain of coupled two-level systems connecting two thermal reservoirs. Through an analytic treatment we find that the energy current is independent of the system size, hence violating Fourier's law of heat conduction. The classical diffusive behavior in Fourier's law of heat conduction can be recovered by introducing decoherence to the quantum systems constituting the chain. Implications of these results on energy transfer in biological light harvesting systems, and the role of quantum coherences and entanglement are discussed.

In recent years, energy propagation in systems that must be described in a quantum mechanical way has become a growing field. This growth is partially due to the fact that the understanding of how energy flow can be controlled and efficiently distributed has been identified as one of the crucial fields of study for the development of modern societies [1, 2]. One of the conceptual pillars in energy transport, the validity of Fourier's law of heat conduction, has become an active area of investigation and has been investigated in classical [3, 4] and quantum systems [5–7].

Since experimental evidence for quantum coherent excitation transport in the early light-harvesting step of photosynthesis has been presented [8, 9], investigations in systems of molecular biology have focused on the question, to what extent quantum mechanics contributes to the near perfect transport efficiency in light-harvesting. The emphasis has been put on the *transient* transport efficiency of an initial excitation in the presence of noise and disorder [10–12]. The experiments have been performed with pulsed femtosecond laser sources to excite and probe the molecule samples, whereas it has been suggested [14–16] that the light-harvesting process *in vivo* would be described more accurately in a steady-state scenario, because the light flux coming from the sun is essentially static on time scales that are relevant for molecular excitation transport. Here, we adopt the steady-state view on excitation transport in photosynthetic light-harvesting complexes, and evaluate the impact of Fourier's law on transport in quantum systems of this particular kind.

An important step in the understanding how Fourier's law emerges from the quantum domain has been done by Michel et al. [6]. In this work Fourier's law is derived for a model system that is a chain of N identical coupled subunits, where each of the subunits has a single ground state and a narrow "band" of equally spaced
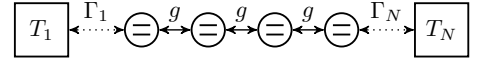
FIG. 1. Chain of two-level quantum systems with its terminal sites coupled to heat baths of different temperature.

excited states. In the present work, we employ a similar system, i.e. a one-dimensional chain of two-level systems, for which we compare the energy current in the classical analogue, where Fourier's law applies, with the quantum case, where we find the energy current to be independent of the chain length. This means that for the one-dimensional chain of two-level atoms Fourier's law applies for the classical variant but there is a distinct violation in the quantum transport scenario. By introducing dephasing to the quantum model, we can study the transition from coherent to incoherent transport and show how Fourier's law can be recovered from the quantum case.

Fourier's law of heat conduction states that the heat current through a classical macroscopic object is proportional to the applied temperature gradient [17],

$$\mathbf{J} = -\kappa \boldsymbol{\nabla} T, \tag{1}$$

where κ is the thermal conductivity. For a one-dimensional homogeneous object, the heat current is therefore determined by the temperature difference of the two heat baths ΔT, and the object length L. Generally, the validity of Fourier's law does not seem to be strictly linked to the classical or quantum nature of the system. For example, in the classical limit, for diffusive systems Fourier's law can be applied, but for ballistic systems in one and two dimensions there are divergences of the thermal conductivity as $\kappa \sim L^{\alpha}$ (see [4] for a review of heat transfer in low dimensional systems). For a discretized object composed of N equally spaced parts (sites), $L \propto N$ and thus

$$J = -\kappa \frac{\Delta T}{L} = -cN^{\alpha} \frac{\Delta T}{N} = -c\,\Delta T\,N^{\alpha-1}, \tag{2}$$

where c is a constant of proportionality. For some one-dimensional quantum systems, one the other hand, there is evidence that Fourier's law is valid, i.e. $\alpha = 0$ [5, 6].

The quantum system considered is a one-dimensional chain of $N \geq 2$ two-level systems with coherent next-neighbor couplings as depicted in Fig. 1. The Hamiltonian is

$$H = \sum_{k=1}^{N} \frac{\hbar\omega}{2} \sigma_k^z + \sum_{k=1}^{N-1} \hbar g \left(\sigma_k^+ \sigma_{k+1}^- + \sigma_k^- \sigma_{k+1}^+ \right), \quad (3)$$

where σ_k^z, σ_k^+, and σ_k^- are the Pauli-z, raising, and lowering operators in the basis of ground and excited state of the kth two-level system, respectively, with on-site energy $\hbar\omega$ and coupling strength g. Similar simple models of coupled effective two-level systems are used in recent analyses of energy transfer in photosynthetic complexes [10–12]. The influence of the two heat baths is modeled by incoherently coupling each of the terminal sites to a bosonic heat bath described by a master equation of Lindblad form. The system dynamics is then described by the master equation

$$\dot{\rho} = -\frac{i}{\hbar}[H, \rho] + \mathcal{L}_1 \rho + \mathcal{L}_N \rho, \quad (4)$$

where $\mathcal{L}_k$ acts on the first (last) site for $k = 1(N)$, respectively, and is given by

$$\mathcal{L}_k \rho = \Gamma_k(n_k + 1) \left(\sigma_k^- \rho \sigma_k^+ - \frac{1}{2} \left\{ \sigma_k^+ \sigma_k^-, \rho \right\} \right)$$
$$+ \Gamma_k n_k \left(\sigma_k^+ \rho \sigma_k^- - \frac{1}{2} \left\{ \sigma_k^- \sigma_k^+, \rho \right\} \right). \quad (5)$$

The first term in $\mathcal{L}_k$ accounts for emission into the reservoir, the second term accounts for absorption, Γ_k is the interaction rate, and $n_k = 1/[\exp(\hbar\omega/(k_B T_k)) - 1]$ is the temperature-dependent mean excitation number at the resonance frequency in the respective bosonic thermal reservoir [13], with k_B being Boltzmann's constant.

The expression of the heat current for a quantum system, J_Q, is derived from the time-derivative of the energy of the system,

$$\dot{E} = \frac{d}{dt} \langle H \rangle = \text{Tr} \left(H \dot{\rho} \right) = 0, \quad (6)$$

which vanishes in the steady state. When inserting (4) into this expression, we obtain

$$0 = \text{Tr} \left(H \mathcal{L}_1 \rho + H \mathcal{L}_N \rho \right) =: J_1 + J_N, \quad (7)$$

on the basis of which one can define the heat current to/from the respective reservoirs, both being of opposite sign, but equal in magnitude [13]. The heat current through the chain is therefore equal to the net energy that enters the network from one reservoir and exits to the other per unit time, i.e. the quantity $J_Q = |J_1| = |J_N|$.

A straightforward evaluation of J_Q for our system in the steady state yields the compact expression

$$J_Q = \gamma_1 \hbar\omega \left(s_1 - \langle \sigma_1^+ \sigma_1^- \rangle \right) - \frac{\gamma_1 \hbar g}{2} \left(\langle \sigma_1^+ \sigma_2^- \rangle + \langle \sigma_1^- \sigma_2^+ \rangle \right), \quad (8)$$

where $\gamma_1 = \Gamma_1(2n_1 + 1)$ denotes the effective coupling to the reservoir, $s_1 = n_1/(2n_1 + 1)$ is the excited-state population of a single two-level system in thermal equilibrium with reservoir 1, and all expectation values are taken with respect to the steady state of the chain. The heat current in the steady state is thus solely characterized by the excited-state population of the first site and its specific energy gap, and since $\langle \sigma_1^+ \sigma_2^- \rangle = \langle \sigma_1^- \sigma_2^+ \rangle^*$, it is furthermore given by the *real* part of the coherence between sites one and two. An analogous expression can be given for the last site of the chain, which is connected to the second heat bath.

For the complete expression of the heat current, we need the excited-state population of the first site, $\langle \sigma_1^+ \sigma_1^- \rangle$, and the coherences between the first two sites, $\langle \sigma_1^+ \sigma_2^- \rangle$. The excited-state populations of the individual sites in the steady state can be obtained from considering specific matrix elements of the master equation of the kind $\frac{\partial}{\partial t} \langle \sigma_k^+ \sigma_k^- \rangle = \text{Tr}(\sigma_k^+ \sigma_k^- \dot{\rho}) = 0$. There are different cases: sites 1 and N, which are connected to their respective heat bath, and the remaining sites, which are in the middle of the chain. The relevant equations for the terminal sites $k = 1$ and $k = N$ yield:

$$\gamma_1 \left(s_1 - \langle \sigma_1^+ \sigma_1^- \rangle \right) = ig \left(\langle \sigma_1^+ \sigma_2^- \rangle - \langle \sigma_1^- \sigma_2^+ \rangle \right),$$
$$\gamma_N \left(s_N - \langle \sigma_N^+ \sigma_N^- \rangle \right) = -ig \left(\langle \sigma_{N-1}^+ \sigma_N^- \rangle - \langle \sigma_{N-1}^- \sigma_N^+ \rangle \right).$$

For the inner sites, $1 < k < N$, we obtain

$$\langle \sigma_{k-1}^+ \sigma_k^- \rangle - \langle \sigma_{k-1}^- \sigma_k^+ \rangle = \langle \sigma_k^+ \sigma_{k+1}^- \rangle - \langle \sigma_k^- \sigma_{k+1}^+ \rangle, \quad (9)$$

that is, the *imaginary* part of all coherences between neighboring sites are equal. These equations motivate the following general form for the excited-state populations of the terminal sites:

$$\langle \sigma_1^+ \sigma_1^- \rangle = s_1 - \Delta/\gamma_1, \quad \langle \sigma_N^+ \sigma_N^- \rangle = s_N + \Delta/\gamma_N. \quad (10)$$

The transport along the chain thus causes a shift of the excited-state population of the terminal sites from the thermal equilibrium by Δ/γ_k, where $\Delta = ig \left(\langle \sigma_1^+ \sigma_2^- \rangle - \langle \sigma_1^- \sigma_2^+ \rangle \right)$. The coherences, and thereby Δ, can be obtained by a similar argument. Summing up coherences of the steady state, $\frac{\partial}{\partial t} \sum_{k=1}^{N-1} \langle \sigma_k^+ \sigma_{k+1}^- \rangle = 0$, provides the equation

$$-ig \left(\langle \sigma_1^+ \sigma_1^- \rangle - \langle \sigma_N^+ \sigma_N^- \rangle \right) = \frac{\gamma_1}{2} \langle \sigma_1^+ \sigma_2^- \rangle + \frac{\gamma_N}{2} \langle \sigma_{N-1}^+ \sigma_N^- \rangle,$$

the imaginary part of which, using (9) and (10), yields

$$\Delta = \frac{4g^2 \gamma_1 \gamma_N (s_1 - s_N)}{(\gamma_1 + \gamma_N)(4g^2 + \gamma_1 \gamma_N)}. \quad (11)$$

Next, for the complete solution of the heat current, we need the real part of the next-neighbor coherences, i.e.
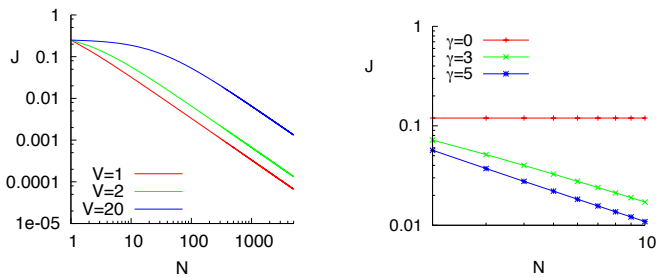
FIG. 2. (Color online) Heat current as a function of the system size in a log-log plot (left) for the classical model for different values of the coupling V, and (right) for the quantum model for different values of the dephasing rate γ. Unless otherwise indicated $V = 1$, $g = 1$, $k_B T_1 = 1$, $\hbar\omega = 1$, $\Gamma_1 = \Gamma_N = 1$, and $T_N = 0$.

$\langle\sigma_1^+\sigma_2^-\rangle + \langle\sigma_1^-\sigma_2^+\rangle$. For a chain of sites with uniform on-site energy as considered here, these coherences are purely imaginary, which can be shown from the structure of the master equation (see appendix). We thus arrive at the final expression of the heat current for a uniform quantum chain

$$J_Q = \hbar\omega\Delta = -2\hbar\omega g \, \mathrm{Im}\langle\sigma_1^+\sigma_2^-\rangle. \qquad (12)$$

Here, we observe an important property of the heat current. It is independent of the chain length and thus violates Fourier's law, that is, the thermal conductivity scales as $\kappa \sim N$, and thus $\alpha = 1$.

If one considers technical applications, this result may open a new perspective to design materials with a more efficient heat transfer, free from losses due to the system size. Furthermore, provided that energy transport in light-harvesting complexes is governed by steady-state properties, this fact also provides a novel perspective on the understanding of highly efficient energy transport.

Next, for comparison of the quantum model with the analogous classical model, we derive the heat current for the latter, which corresponds to the symmetric simple exclusion process [18], or Förster-type hopping [19]. It is a chain of N sites, which each may carry a single particle (excitation) that probabilistically moves between neighboring sites. This diffusive model fulfills Fourier's law. The classical probability for a particle to be at site k is given by P_k. The master equation (4) is thus turned into a Pauli master equation, i.e. a set of classical rate equations:

$$\dot{P}_1 = \Gamma_1 n_1 + P_1\big(-\Gamma_1(n_1+1) - \Gamma_1 n_1 - V\big) + V P_2$$
$$\dot{P}_k = V\left(P_{k-1} + P_{k+1} - 2P_k\right) \qquad (k \neq 1, N)$$
$$\dot{P}_N = \Gamma_N n_N + P_N\big(-\Gamma_N(n_N+1) - \Gamma_N n_N - V\big) + V P_{N-1}$$

where V is the constant rate to hop between sites. In the classical system, the heat current is defined by $J_C = |V\left(P_{i+1} - P_i\right)|$, i.e. the net transfer rate of energy between sites, which in the steady state yields

$$J_C = \frac{\gamma_1\gamma_N V (s_1 - s_N)}{V(\gamma_1 + \gamma_N) + \gamma_1\gamma_N(N-1)}.$$
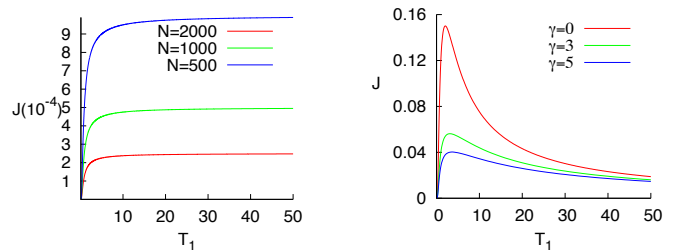

FIG. 3. (Color online) Heat current as a function of temperature T_1 with $T_N = 0$ fixed, for the classical transport for different sizes of the system (left), and the quantum transport for $N = 4$ and different dephasing ratios (right). Parameters are $k_B = 1$, $\hbar\omega = 1$, $\Gamma_1 = \Gamma_N = 1$, $V = 1$, and $g = 1$.

In the limit $N \to \infty$, the heat current scales with the system size as $J_C \sim V(s_1 - s_N)/N$. Therefore, the heat current of the classical analogue obeys Fourier's law with $\kappa = \mathrm{const.}$ and $\alpha = 0$, in contrast to the quantum system.

The difference between the heat current of quantum and classical systems can be lifted by adding a dephasing environment to each site of the quantum model. This amounts to introducing an additional term for every site in the master equation (4):

$$\mathcal{L}_{\mathrm{deph}}\rho = \gamma\sum_{k=1}^N\left(\sigma_k^+\sigma_k^-\rho\sigma_k^+\sigma_k^- - \frac{1}{2}\left\{\sigma_k^+\sigma_k^-, \rho\right\}\right). \quad (13)$$

The results for the classical and quantum chain are given in fig. 2 on a log-log scale. The classical model features a linear dependence in the system size, for high enough values of N as expected. The heat current of the quantum case without dephasing is also linear in N, but *constant*. However, when additional dephasing is applied, the heat current is suppressed and now features a size-dependence as $1/N$ for sufficiently large values of the dephasing rate γ, as confirmed by a numerical analysis of fitting the heat current to a power-law (see appendix). By adding dephasing to the quantum system, we can thus recover the classical $1/N$-dependence of the heat current.

In a common interpretation of a dephasing environment, dephasing is caused by fluctuations of the on-site energy of every site. The excited state then effectively forms a band of states that is separated by a gap from the ground state. Adding dephasing thus effectively recovers the quantum model treated in [6], and yields the same qualitative result concerning the validity of Fourier's law regarding its dependence on the system size.

Turning from the system size to the temperature-dependence, we find that in the quantum system the heat current features a strong dependence on the temperatures of the heat baths. Fig. 3 collects the temperature-dependencies for both models. The heat current of the classical system saturates for high values of the temperature, which constitutes a violation of Fourier's law. This is due to the fact that the system has only two levels, which implies a finite heat capacity of the system. Therefore, it cannot transport an arbitrary large amount of
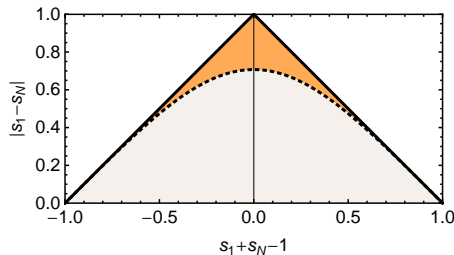
FIG. 4. Region of parameter space where the steady state may exhibit entanglement for $\gamma_1 = \gamma_N$. Within the shaded area of all possible values for s_1 and s_N, the upper (darker) parameter region indicates values for s_1 and s_N where entanglement can occur (dashed boundary not included). That is, only for s_1 and s_N in this region, parameters g and $\gamma_1 = \gamma_N$ exist such that the steady state is entangled. Entanglement cannot occur for any values of the coupling parameters outside the darker shaded region.

energy, and thus cannot scale linearly with the temperature for a large temperature difference. The quantum transport features a more intricate behavior. For a high temperature of the hot heat bath, its mean number of excitations and thereby γ_1 increases, causing a Zeno-type effect that reduces the transport efficiency of the system. With additional dephasing, the temperature-dependence of the heat current of the quantum system approaches a qualitatively similar saturating behavior as in the classical system.

Considering excitation transfer processes in light-harvesting complexes of photosynthesis, there are several lessons to be drawn from our model. Assume that, individual sites of the chain represent the pigments that can carry an excitation, e.g. chlorophyl molecules, and the hot and cold reservoirs, which supply and consume excitation, correspond to the radiation field and photosynthetic reaction centers, respectively. Even though the coupling topology of the network of pigments is not necessarily linear, we can arrive at a number of principle observations: There exist quantum coherences in the system in the steady state that are static. It is these coherences alone that determine the transport properties as indicated in (12). Secondly, the violation of Fourier's law for the quantum transport provides a new way of how Nature may exploit quantum mechanics to gain size-independence, and thus for larger complexes to achieve a more efficient excitation throughput.

A relevant point in this respect is the influence of disorder in the system, and the observation that additional noise may unlock the effect of localization in disordered systems for transient transport processes [10, 11]. However, figs. 2 and 3 show that additional noise due to local dephasing reduces the observed heat current. Although, here, this result is obtained for a chain with uniform on-site energies and inter-site couplings, i.e. in the absence

of disorder, we have also numerically investigated disordered chains. To this extent we have sampled the heat current in chains with $N = 5$, with all on-site energies $\hbar\omega_k$ and couplings g_{kl} randomly chosen from a uniform distribution in the interval $[0, 1]$. In 8662 of the 10 000 disorder samples, we found dephasing to reduce the heat current. Whenever additional dephasing is found to increase the heat current, the original random configuration exhibited a heat current below average of the entire random ensemble. We thereby extended what has been observed in the transient case [12] to the steady-state scenario.

With the perspective of identifying conceivable biological realizations of this transport scenario, an interesting aspect is the question whether entanglement is generated and what role it plays, as addressed in [20, 21]. Fig. 4 summarizes for which parameters entanglement of the non-equilibrium steady state occurs for a chain with $N = 2$ and equal effective bath rates $\gamma_1 = \gamma_N$. We find that entanglement can occur, but only in specific regions of the parameter space. Furthermore, for rates $\Gamma_1 = \Gamma_N$, the steady state is never entangled for any choice of bath temperatures and coupling g. A bias in the bath rates, however, may drive the system to an entangled steady state. Depending on the interaction strength between the sites, entanglement may exist for certain range of temperatures. In contrast to entanglement studies in photosynthesis [21], in the present scenario we find that the occurrence of entanglement is not equivalent to, and does not necessarily come with the mere presence of coherences. It is thus an additional feature.

To conclude, we have analyzed the energy transfer in a quantum system, formed by a paradigmatic chain of two-level systems, for which we found the heat current in the steady state to be independent of the chain length. This finding constitutes an important addition and gives a new twist to the study of the validity and emergence of Fourier's law in classical and quantum systems. We recover Fourier's law in the quantum-to-classical transition by adding dephasing that destroys quantum coherences. On the other hand, the violation of Fourier's law in the considered system encourages applications of quantum systems for efficient energy transport in specially designed materials by exploiting the size-independence. From the viewpoint of (excitation) energy transport in biological systems, the violation of Fourier's law constitutes another possibility of how quantum effects may be exploited in light-harvesting systems. It is the coherences in the system that govern the transport properties by design, whereas entanglement may appear independently and in addition for a sufficiently large non-equilibrium.

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I. APPENDIX

A. Purely imaginary coherences

From the structure of the master equation in Lindblad form (4), an ordinary linear differential equation, one can directly infer that next-neighbor coherences are imaginary. In Liouville space the equation reads $\dot{\rho} = L\rho$, where ρ is the vector of all matrix elements, which are coupled linearly by the matrix L, the Liouvillian. For our purposes, it is helpful to introduce notation for the matrix elements:

$$_{12\ldots N}\langle ik\ldots q|\rho|jl\ldots r\rangle_{12\ldots N} \equiv \rho_{ij,kl,\ldots,qr},$$

where indices are grouped by subsystem. Vectors are products of basis vectors of the individual sites with ground state $|0\rangle$ and excited state $|1\rangle$. We thus treat matrix elements with possible indices "0" and "1".

The Liouvllian L is a sum of three parts, each of which couples certain matrix elements, which yields independent sets of coupled matrix elements. It is possible to distinguish independent sets by observing general rules of how the Liouvillian couples matrix elements. We formulate these rules by the way indices are transformed by the Liouvillian.

The Lindblad terms $\mathcal{L}_k$ of the Liouvillian inject or extract excitations at the terminal sites of the chain. Thereby, they transform matrix elements into one another that differ only by a pair of "00" and "11" indices of the first/last subsystem, e.g. $\rho_{\mathbf{00},01} \leftrightarrow \rho_{\mathbf{11},01}$. This constitutes a change of the total number of indices "0" and "1" by two, hence leaving the respective total number of indices "0" and "1" even or odd. Since H commutes with the excitation number operator, the commutator that appears in the Liouvillian leaves the total number of excitations invariant and hence couples only matrix elements with the same number of indices "0" and "1", respectively. The coherent dynamics captures the exchange of excitations between neighboring sites and, in terms of matrix elements, couples those that can be transformed into each other by exchanging a "0" and a "1"-index between neighbors, while maintaing the relative index position, i.e. left and right indices are transformed within themselves, e.g. $\rho_{\mathbf{01},\mathbf{10}} \leftrightarrow \rho_{\mathbf{11},\mathbf{00}}$. This implies that the ground state $\rho_{00,00,\ldots}$ is coupled to all other populations, i.e. matrix elements with indices of the form $\rho_{ii,jj,\ldots}$, and only to those coherences that contain an equal number of indices "1" on the left and right. The remaining matrix elements form an independent closed set of equations, whose steady-state solution is therefore the trivial solution, where all matrix elements vanish. (The set of equations that includes the populations is not solved by the trivial solution in the steady-state because it is subject to the boundary condition $\mathrm{Tr}\,\rho = 1$.) Note, that the diagonal of L contains only coefficients with negative real

parts, meaning that all matrix elements would decay to zero if not sufficiently maintained by a positive contribution due to another element. A population is coupled to a next-neighbor coherence, e.g. $\rho_{11,00} \leftrightarrow \rho_{01,10}$, with a coupling $\pm ig$ such that (real) populations pump the imaginary part of the next-neighbor coherences, and vice versa. The contributions to/from the real part of the latter cancel, leading to their decay. In longer chains ($N > 2$) next-neighbor coherences are also coupled to next-to-nearest neighbor coherences, e.g. $\rho_{01,10,00} \leftrightarrow \rho_{01,00,10}$, with the same factor $\pm ig$ thus coupling imaginary (real) part of the former to real (imaginary) part of the latter. Therefore, imaginary and real parts of next-neighbor coherences belong again to different and independent sets of coupled differential equations. The real parts have the trivial solution, whereas the imaginary part is non-zero in the steady state.

B. Fit of dephasing numerics

To analyze the behavior of the quantum system under the effect of dephasing, we take the logarithm of (2) and obtain a linear dependence between $\log J$ and $\log N$.

$$\log J = \log\left(c\Delta T\right) + (\alpha - 1)\log N.$$

By a linear regression of the numerical data of fig. 2, for $\gamma = 5$ we obtain a value $\alpha = 0.0242$ with a regression coefficient $R = 0.9999851$. The small discrepancy with Fourier's law ($\alpha = 0$) is due to the finite-size of the system.